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Dielectric Behaviour of CCTO-Silicone Resin Composites in 0-3 Connectivity

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ABSTRACT

The dielectric behaviour of CCTO-silicone resin composites was analyzed as a function of ceramic amount. Composites were prepared by mixing the components and pouring them in to suitable moulds. Dielectric measurements were performed from 10Hz to 1MHz and 30 to 200^0C. It was observed that both dielectric constant (ε) and loss tangent (tan δ) increased gradually with the increase in CCTO content. For the composite with CCTO volume fraction of 0.9, the dielectric constant and loss tangent at 1 kHz was 151 and 0.35, respectively. In comparing with pure silicone resin (ε = 3.0 and tan δ = 0.03), the dielectric constant (ε) of the composite was improved by 50 times, while the loss tangent (tan δ) increased nearly 12 times. High ε and low tan δ of the CCTO-silicone resin composites made it attractive for practical applications. Various theoretical models were employed to predict the dielectric constant of these composites, the dielectric constant obtained via Maxwell-Garnett model was in close agreement with the experimental data.

Keywords: CCTO, silicone resin, composites, dielectric constant and loss tangent.

1. INTRODUCTION

Ceramic-polymer composites with high dielectric constant have been studied widely due to their good fabrication process, low cost and potential applications in many fields such as dielectric films in embedded capacitors and electric energy storage devices[1]. In previous studies, $BaTiO_3$-polymer composites for embedded

capacitors were introduced and fully characterized[2-3]. However, the dielectric constant of such polymer based composites is rather low (~50) because of the lower dielectric constant of the matrix[4]. For instance, in $BaTiO_3$/epoxy composites, though $BaTiO_3$ has relatively high dielectric constant (>1000), the effective dielectric constant of the composite was as low as 50, even when the highest possible volume fraction of ceramics was incorporated. As the volume fraction of ceramics increased, the composite, unfortunately, lost its flexibility. Perovskite-like structure $CaCu_3Ti_4O_{12}$ (CCTO) dielectric ceramic has been paid much attention in the field of materials science and application for its colossal dielectric constant, independence of dielectric properties of the frequency and temperature in a wide range and nonlinear behaviours[5]. A number of theoretical studies and experimental observations have attempted to elucidate the remarkable dielectric properties of CCTO perovskite-like material. This material has demonstrated to have a dielectric constant as high as 50,000. In this work, an attempt has been made to study dielectric behaviour of CCTO-silicone resin composites in 0-3 connectivity.

2. EXPERIMENTAL

CCTO: silicone resin composites of 0-3 connectivity were prepared by mixing pre sintered powder of CCTO ceramic. The first set of samples were prepared in such a way that the material contains 90 percent (90%) by volume of CCTO ceramic and 10 percent (10%) by volume of silicone resin. A paste of the ceramic and resin is formed; one assumes that the CCTO powder has been evenly distributed into a matrix of silicone resin. To 0.5 % by weight of the prepared paste, dibenzoyl peroxide was added and the paste was again mixed so that the peroxide distributes evenly throughout the volume of the mixture. Dibenzoyl peroxide acts as a cross linking agent between the resin molecules. The paste is now injected into a steel die (mould) and the mould loaded with the paste is then heated to 140^{o}C. The temperature was held for 30 min after which the heater was turned off and the mould was allowed to cool to room temperature, opened and the material inside the cavity is removed. We obtained a cured sample which is rubber like, since the silicone resin now acts like an elastic solid, with CCTO ceramic powder distributed within the matrix like filler. This is now a 0:3 ceramic polymer composite samples. The resulting sample yields a thickness of not more than 1.5 mm. The procedure mentioned above was repeated for samples of compositions 80%, 70%, 60% and 50% by volume of CCTO. The dielectric properties of the sample were determined using the HP 4192A LF Impedance Analyzer.

3. RESULTS AND DISCUSSION

Figure 1 illustrates the dielectric constant (ε) and the loss tangent (tan δ) of the composites measured at 1 kHz and room temperature. It was found that both ε and tan δ increased gradually with the increase in CCTO content. For the composite with CCTO volume fraction of 0.9, the ε and tan δ values at 1 kHz were 151 and 0.35, respectively. In comparing with pure silicone resin (the experimental values of pure silicone resin, ε = 3.0 and tan δ = 0.03),

the ε value of the composite was improved by 50 times, while the tan δ value increased by nearly 12 times. High ε and low tan δ of the CCTO-silicone resin composite made it attractive for practical applications. Figure 2 and 3 show dielectric constant as a function of frequency and temperature, respectively. As it can be expected, dielectric constant rose as the ceramic volume fraction increases. In order to study the frequency and temperature dependence of relaxation processes, electrical modulus was used. Figure 4 show the real and imaginary parts of the electrical modulus obtained through equation (1) as a function of frequency and temperature, respectively.

$$M^* = \frac{1}{\varepsilon^*} = \frac{1}{\varepsilon' - \varepsilon''} = \frac{\varepsilon'}{\varepsilon'^2 + \varepsilon''^2} + j\frac{\varepsilon''}{\varepsilon'^2 + \varepsilon''^2} = M' + jM'' \tag{1}$$

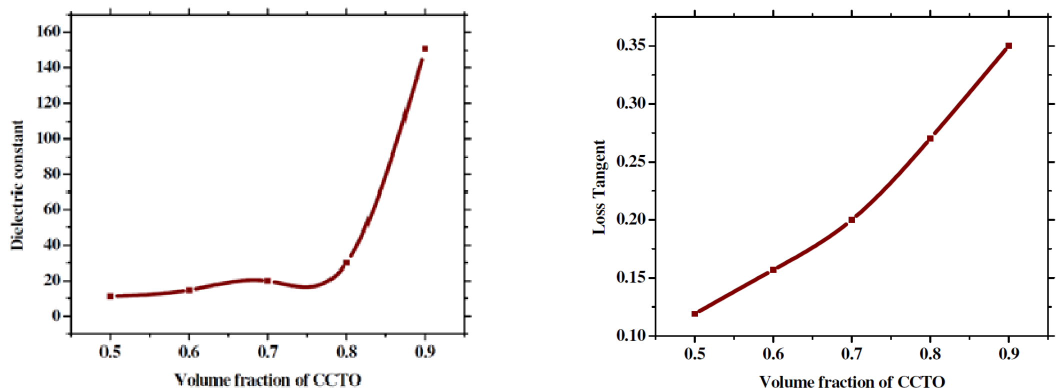

Figure-1: Dependence of dielectric constant and loss tangent of the CCTO-silicone resin composites on the volume fraction of CCTO at 1 kHz and room temperature.

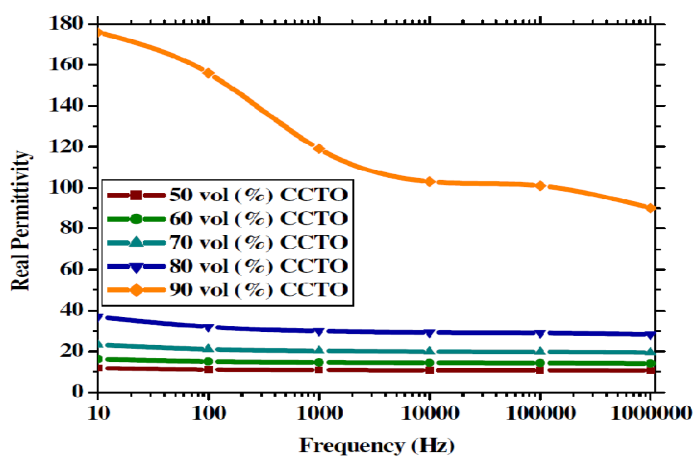

Figure-2: Dielectric constant (Real Permittivity) vs. frequency for composites with different CCTO volume fraction at temperature 30 °C.

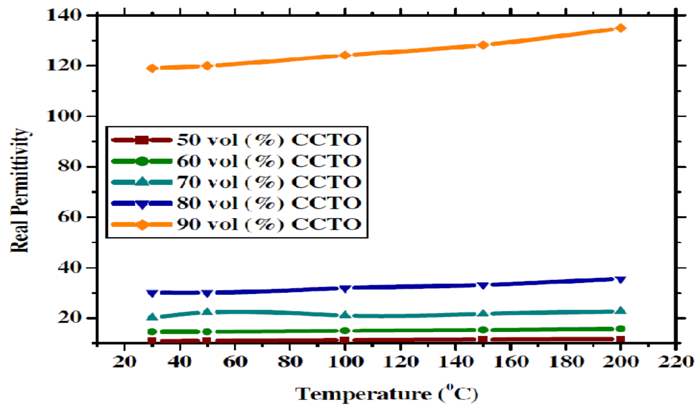

Figure-3: Dielectric constant (Real Permittivity) vs. temperature for composites with different CCTO volume fraction at frequency 1000 Hz.

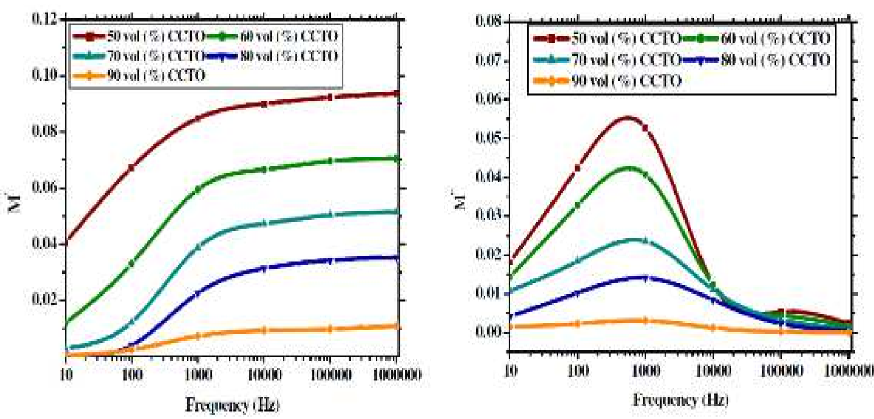

Figure-4: Real part (M') and imaginary part (M'') of electrical modulus vs. frequency and volume CCTO fraction at 100 ^{0}C.

In figure 4, it can be seen that M´ values increased with frequency. Nevertheless, peaks in M´´ values were developed at this same frequency range, indicating the appearance of a relaxation process (α relaxation). The maximum of M´´ decreased when filler amount increased. Relaxations peaks were displaced to higher frequencies, since relaxation processes were influenced by the interfacial polarization effect which generated electric charge accumulation around the ceramic particles and the displacement of peak as the particle content increased. A number of numerical relations have been put forward by researchers to predict the dielectric constant of the composite. In this work, the following equations have been used to calculate the effective dielectric constant of the CCTO-silicone resin composites.

$$log\varepsilon_C = V_m log\varepsilon_m + (0.7)V_f log\varepsilon_f \tag{2}$$

$$\varepsilon_C = \varepsilon_m[1 + 3V_f\{\frac{(\varepsilon_f - \varepsilon_m)}{(\varepsilon_f + 2\varepsilon_m)}\}] \tag{3}$$

$$\varepsilon_C = \varepsilon_f[1 + \{3V_f\,(\varepsilon_m - \varepsilon_f)\}/\{2\varepsilon_f + \varepsilon_m - V_f(\varepsilon_m - \varepsilon_f)\}] \quad (4)$$

Equations (2)-(4) are the expressions of Lichtenecker, Clausius Mossotti and Maxwell-Garnett models respectively, where ε_C, ε_f and ε_m are the dielectric constants of the composites; CCTO ceramic powder and silicone resin respectively and V_f and V_m are the volume fractions of the CCTO ceramic and silicone resin respectively. The dielectric constants of silicone resin and CCTO ceramic measured at 1 kHz are 3.0 and 139, respectively.

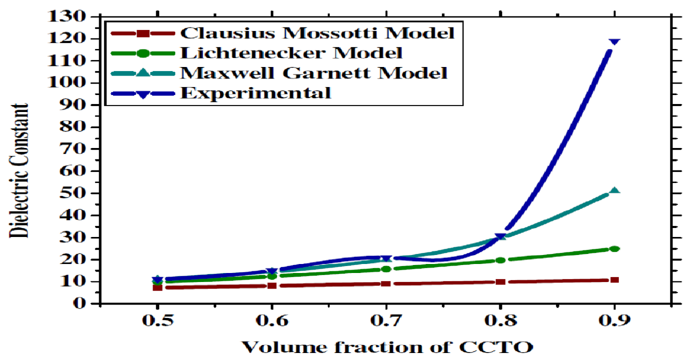

Figure-5: Comparison of experimental and theoretical dielectric constants of CCTO-silicone resin composites at 1 kHz.

Figure 5 depicts the comparisons of dielectric constants of the CCTO-silicone resin composites with the values predicted by the above equations at 1 kHz. From this figure, it is clear that the Maxwell-Garnett model is the best fit of the experimental values for the CCTO-silicone resin composites. Maxwell-Garnett model predicts that the dielectric constant of the composite is nearly equal to the experimental values up to 0.8 CCTO volume fractions. However, as volume fraction of CCTO increases beyond 0.8, a deviation from the predicted value of dielectric constant is observed in CCTO-silicone resin composites.

4. CONCLUSIONS

$CaCu_3Ti_4O_{12}$ (CCTO) ceramic-silicone resin polymer composite in 0-3 connectivity fabricated by using CCTO ceramic particles as filler and silicone resin as polymer matrix is introduced in the present work. The dielectric constant (ε) and the loss tangent (tan δ) of the composites measured at 1 kHz and room temperature were increased gradually with the increase in CCTO content. For the composite with CCTO volume fraction of 0.9, the ε and tan δ at 1 kHz was 151 and 0.35, respectively. In comparing with pure silicone resin (the experimental values of pure silicone resin, ε = 3.0 and tan δ = 0.03), the ε of the composite was improved by 50 times, while the tan δ increased nearly 12 times. In order to study the frequency and temperature dependence of relaxation processes, electrical modulus was used. The dielectric constant of the composite was simulated based on three different models. The values

obtained by the Maxwell-Garnett model are in close agreement with the experimental values up to 0.8 CCTO volume fractions.

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